

THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of	)	
Mikio HASHIMOTO et al.	)	Group Art Unit: 1713
Application No.: 09/973,902	)	Examiner: Donald R. Wilson
Filed: October 11, 2001	)	Appeal No.:
For: PROPYLENE POLYMER	)	
COMPOSITIONS	)	

**BRIEF FOR APPELLANT**

**Mail Stop APPEAL BRIEF - PATENTS**

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

This appeal is from the decision of the Primary Examiner dated June 10, 2003 (Paper No.6) finally rejecting claims 1, 2 and 4-17, which are reproduced as an Appendix to this brief.

A check covering the ☐ \$165.00 (2402) ☒ \$330.00 (1402) Government fee and two extra copies of this brief are being filed herewith.

The Commissioner is hereby authorized to charge any appropriate fees under 37 C.F.R. §§1.16, 1.17, and 1.21 that may be required by this paper, and to credit any overpayment, to Deposit Account No. 02-4800. A copy of this page and the signature page are submitted in triplicate.

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I. Real Party in Interest

The present application is assigned to Mitsui Chemicals, Inc..

II. Related Appeals and Interferences

The Appellant's / Appellants' legal representative, or assignee, does not know of any other appeals or interferences which will affect or be directly affected by or have bearing on the Board's decision in the pending appeal.

III. Status of Claims

Claims 1-17 are currently pending in this application. Claim 3 stands allowed.

IV. Status of Amendments

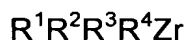
There are no unentered amendments in this application.

V. Summary of the Invention

The claims on appeal are directed to propylene polymer compositions comprising blends of propylene polymers, the individual polymers in the blends being prepared by polymerizing propylene monomer using specified catalysts. The compositions may be produced by either separately preparing the individual polypropylenes and blending them together, as in claim 1, or sequentially preparing the blend *in situ* in a multi-stage polymerization process by polymerizing propylene under a first set of conditions to obtain a first polymer and subsequently polymerizing propylene under a second set of conditions to obtain a second polymer whereby the blend is produced in one or plural reactors (see claim 2). Note pages 35 and 36 of the specification.

Specifically, the first propylene polymer in the blends of the invention is prepared by polymerizing a monomer comprising propylene using a catalyst combination comprising:

- (1) a zirconocene compound of the formula



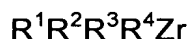
where two of the substituents  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  are indenyl groups substituted with an aryl group and linked through a dimethylsilylene and the other two substituents  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  are halogen atoms, and

- (2) at least one organoaluminum oxy-compound.

The polymerization conditions are manipulated to obtain a first polypropylene having a melt flow rate (MFR) of 0.01 to 30 g/10 min. and a molecular weight distribution (Mw/Mn) of 2 to 3. Note the discussion on pages 32-33 of the specification.

The second propylene polymer in the blends of the invention is prepared by polymerizing a monomer comprising propylene using a catalyst combination comprising:

- (1) a zirconocene of the formula



where two of the substituents  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  are indenyl groups substituted with an aryl group and linked through a dimethylsilylene and the other two substituents  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  are halogen, and

- (2) at least one organoaluminum oxy-compound.

The polymerization conditions are such as to yield a second polypropylene having a melt flow rate (MFR) of 30 to 1000 g/10 min. and a molecular weight distribution (Mw/Mn) of 2 to 4. Note page 33 of the specification.

The first and second propylene polymers are present in the blends in an amount of 10 to 90 wt.% of the first polymer and 90 to 10 wt.% of the second polymer. The ratio of the melt flow rate of the second polymer to the melt flow rate of the first polymer is not less than 30. Note page 34 of the specification.

Suitable zirconocene catalysts are listed on pages 41-52 of the specification. Preferred catalysts are described in the specification on pages 53-63. Suitable organoaluminum oxy-compounds are disclosed on pages 65-67.

Another feature of the present invention resides in the addition of a soft polymer to the propylene polymer blends described in claims 1 and 2. The soft polymer comprises a (co)polymer of ethylene or an  $\alpha$ -olefin of 3 to 30 carbon atoms having a melt flow rate of 0.01 to 100 g/10 min. and a crystallinity of less than 30%. The amount of the soft polymer added to the blend ranges from 3 to 30 parts by weight per 100 parts by weight of the blend. The feature is set forth in claims 4 and 5. The soft polymer is discussed in the specification on pages 77-79. Blends containing the soft polymer are described on pages 79-81.

Appellants have discovered that blends of propylene polymers where the propylene polymers are prepared using zirconocene compounds as defined in the claims on appeal (i.e., having aryl-substituted indenyl groups), unexpectedly exhibit heat distortion temperatures which are significantly higher than blends where the propylene polymers are prepared using similar zirconocene compounds containing alkyl-substituted indenyl groups.

## VI. The Issues

The issues raised in this appeal are as follows:

(1) whether claims 1, 2 and 6-17 were properly rejected under 35 U.S.C. §103(a) as obvious over U.S. Patent No. 5,278,264 to *Spaleck et al.* taken with U.S. Patent No. 5,218,052 to *Cohen et al.* and JP 59-172507.

(2) whether claims 4 and 5 were properly rejected under 35 U.S.C. §103(a) as obvious over U.S. Patent No. 5,278,264 to *Spaleck et al.* in view of U.S. Patent No. 5,218,052 to *Cohen et al.* and JP 59-172507, and further in view of EP 0 518 125, U.S. Patent No. 4,251,646 to *Smith et al.*, Exxon (article by *Yu et al.* entitled "Polyolefin Modification with EXACT™ Plastomers"), U.S. Patent No. 4,634,735 to *Thiersault et al.* and/or EP 0 294 770.

VII. Grouping of Claims

Claims 1, 2 and 12 are independent claims. For purposes of this appeal, claims 1, 2, 6, 7, 9, 10 and 12-17 stand or fall together.

Independent claim 3 stands allowed. A copy of this claim is reproduced in Appendix B. Claims 8 and 11 are directly dependent on claim 3. Thus, claims 8 and 11 should be grouped separately from claims 1, 2, 6, 7, 9, 10 and 12-17.

Claims 4 and 5 are separately patentable from claims 1, 2 and 6-17 and should be grouped separately.

VIII. Arguments

A. Rejection of claims 1, 2 and 6-17

In the first action final rejection mailed June 10, 2003, claims 1, 2 and 6-17 were rejected under 35 U.S.C. §103(a) as obvious over *Spaleck et al.* '264 taken with *Cohen et al.* '052 and JP '507. According to the Examiner, the primary reference discloses polymerizing propylene using a metallocene catalyst which "can have a formula within the scope of applicant's claimed formula." The Office Action acknowledges that *Spaleck et al.* '264 does not disclose blends of propylene polymers of higher and lower molecular weights. The secondary art is relied upon to support the Examiner's allegation that "the use of mixtures of higher and lower molecular weight polypropylene polymers to obtain a broad molecular weight distribution having improved rigidity, moldability and processability, is well known in the art" (page 3, lines 2-4 of the Office Action). The rejection concludes that "it would have been obvious to one of ordinary skill in the art to formulate mixtures of high and low molecular weight polymers with the polymers taught by Spaleck in order to attain the same benefits taught by either JP '507 or Cohen for mixtures of high and low molecular weight polypropylenes."

Initially, Appellants point out that claims 8 and 11 are directly dependent upon allowed independent claim 3. Since claim 3 is novel and unobvious, it necessarily

follows that claims 8 and 11 also are novel and unobvious. Accordingly, the §103(a) rejection of claims 8 and 11 should be reversed.

*Spaleck et al.* '264 discloses a catalyst comprising a metallocene compound and an aluminoxane to prepare olefin polymers having a narrow molecular weight distribution. Although formula I in column 2 of *Spaleck et al.* '264 does generically encompass the zirconocene compounds specified in the claims on appeal, formula I of the reference also encompasses many hundreds of compounds which are outside the scope of the zirconocene compounds of the present claims. The particularly preferred metallocenes disclosed in *Spaleck et al.* '264 are set forth in column 4, last two lines through column 5 (compounds of the formulae A, B and C). The compounds of Formulae A and C do not contain a dimethylsilylene group. None of the particularly preferred compounds of Formulae A, B and C contain aryl-substituted indenyl groups.

It is also noted that the specific metallocene compounds prepared in the working examples of *Spaleck et al.* '264 (identified as Metallocenes A, B, C, D, E, F and G) are different from the zirconocenes defined in the present claims because Metallocenes A-G do not have aryl-substituted indenyl groups. Accordingly, there is no disclosure in the primary reference which would motivate those of ordinary skill in this art to select the zirconocene compounds utilized by Appellants as opposed to any of the other metallocenes generically or specifically disclosed therein. Absent Appellants' own disclosure, only by a fortuitous picking and choosing among many substituents disclosed in the primary reference could one arrive at the zirconocenes employed by Appellants.

As acknowledged in the Office Action, *Spaleck et. al.* '264 contains no disclosure or suggestion of polypropylene blends. Nor does the reference contain any teaching that the polyolefins prepared therein possess improved mechanical properties in comparison to similar polyolefins prepared in the prior art.

*Cohen et al.* '052 discloses a multi-stage polymerization process for preparing polypropylenes having a broadened molecular weight distribution, as opposed to the stated objective of *Spaleck et al.* '264 to prepare polymers of a narrow molecular

weight distribution. The catalysts employed in *Cohen et al.* '052 are not metallocenes, rather they are composites of a titanium-containing component supported on a magnesium-containing compound, combined with a Group II or III metal alkyl, and a silane.

JP '507 discloses a process for polymerizing propylene using a Ziegler-type catalyst. The polymerization takes place in two stages and produces a blend of polypropylenes.

To establish a *prima facie* case of obviousness, three basic criteria must be met. First, there must be some suggestion or motivation, either in the references themselves or in the knowledge generally available to one of ordinary skill in the art, to modify the reference or to combine reference teachings. Second, there must be reasonable expectation of success. Finally, the prior art reference (or references when combined) must reach or suggest all the claim limitations. The motivation to modify the relied on prior art must flow from some teaching in the art that suggests the desirability or incentive to make the modification needed to arrive at the claimed invention. *In re Napier*, 55 F.2d 610,613; 34 U.S.P.Q.2d 1782,1784 (Fed. Cir. 1995). Obviousness cannot be established by modifying the teachings of the prior art to produce the claimed invention, absent some teaching, suggestion or incentive supporting the modification. *In re Geiger*, 815 F.2d 686,688; U.S.P.Q.2d 1276,1278 (Fed. Cir. 1987).

Appellants submit that there is nothing in the secondary references that would provide motivation to those of ordinary skill to vary the process conditions of *Spaleck et al.* '264 to produce a mixture of polypropylenes having a broad molecular weight distribution. Since there is no discussion in *Spaleck et al.* '264 regarding the mechanical properties of the propylene polymers, there would have been no incentive to modify the process disclosed therein to provide polymer blends having improved properties.

The catalytic systems utilized in the secondary references are significantly different from the metallocene catalysts utilized by the primary reference. Those skilled in the art could not reasonably predict that the polymerization system and



conditions described in *Spaleck et al.* '264 could be manipulated such as to prepare a blend of propylene polymer having the characteristics set forth in the appealed claims. The §103(a) rejection represents nothing more than combining isolated disclosures to arrive at Appellants' invention. Since the invention of *Spaleck et al.* '264 is designed to produce polypropylenes having a narrow molecular weight distribution and the processes of the secondary art are designed to produce polypropylenes having a wide molecular weight distribution and in view of the completely different catalytic system used in the references, Appellants respectfully submit that there is no reasonable basis for combining the respective teachings of the cited art.

In a sense, the objective expressed in *Spaleck et al.* '264 to prepare polymers having a narrow molecular weight distribution actually teaches away from the objectives of the secondary art to prepare polymers having a broad molecular weight range. Thus, those of ordinary skill in the art would have been discouraged from modifying the process of *Spaleck et al.* '264 in accordance with the teachings of *Cohen et al.* '052 or JP '507.

For at least the above reasons, Appellants believe that the §103(a) rejection based on *Spaleck et al.* '264 combined with *Cohen et al.* '052 and JP '507 does not establish a *prima facie* case of obviousness. Only impermissible hindsight has been used to arrive at the presently claimed invention.

Assuming, *arguendo*, that a *prima facie* case of obviousness has been established, Appellants submitted a Declaration pursuant to 37 C.F.R. §132 during prosecution to establish the unexpected nature of the present invention. However, the Examiner concluded that the data was unconvincing. Appellants believe the Examiner's position is unsound for at least the following reasons.

The data in the Declaration submitted July 11, 2000 (copy attached as Appendix C) shows that blends of propylene polymers prepared in accordance with the present invention (i.e., using a catalyst including a zirconocene compound having aryl-substituted indenyl groups) have heat distortion temperatures (HDT) significantly higher than that of a comparable blend containing propylene polymers prepared

using a preferred zirconocene compound disclosed in *Spaleck et al.* '264. Specifically, the blends of Exp. 1 and Exp. 2 where both polymers satisfy the conditions of the claims, have very high HDT values of 123 and 121. In contrast, Comp. Exp. 1 and 2 have HDT values of 108 and 109, respectively. This data shows a significant difference which could not have been predicted from the teachings of the cited art and represents unexpected results.

This Declaration was evaluated and discussed in paragraph (4) of the Office Action mailed August 21, 2000. While not disputing the data or the conclusions drawn therefrom, the Examiner argued that the Declaration was unpersuasive because the Declaration did not disclose the procedure used to prepare polypropylenes \*1, \*2 and \*3 nor was the showing in the Declaration commensurate in scope with the claims.

Respectfully, Appellants disagree with the Examiner's position. The polymerization conditions used to prepare the polypropylenes representative of the polymers of *Spaleck et al.* '264 are not considered germane to the issue. The significant issue is that the polypropylenes were prepared using a preferred catalyst disclosed in the primary reference and the propylene polymers had MFR's within the ranges set forth in the present claims. Further, adequate basis exists for reasonably concluding that the zirconocenes encompassed by the present claims would behave in the same manner as the tested catalyst, absent any cogent reason to expect a different result with another catalyst encompassed by the claims.

Appellants respectfully submit that the data provided in the Declaration by Mr. Mori filed July 12, 2000, shows the unexpected results attained by the present invention. The showing effectively rebuts any presumption that the rejected claims are *prima facie* obvious over the combination of *Spaleck et al.* '264 taken with *Cohen et al.* '052 and JP '507.

B. Rejection of claims 4 and 5

Claim 5 is dependent upon allowed claim 3. Since claim 3 is novel and unobvious, it necessarily follows that claim 5 likewise is novel and unobvious. Accordingly, the §103(a) rejection of claim 5 should be reversed.

With respect to the rejection of claim 4, the various tertiary references alone or in combination do not supply the aforementioned deficiencies in the basic §103(a) rejection of claims 1 and 2. Many blends of  $\alpha$ -olefin polymers are known in the art. Those of ordinary skill could not predict with a reasonable degree of certainty what effect the addition of a soft polymer of  $\alpha$ -olefins would have on the properties of a blend of propylene polymers of higher and lower molecular weights prepared in accordance with the combined teachings of *Spaleck et al.* '264, *Cohen et al.* '052 and JP '507.

Accordingly, the combined disclosures of the myriad references relied upon by the Examiner to reject claim 4 fail to overcome the fact that the blends of claim 4 are neither disclosed nor suggested in the cited prior art. For at least these reasons, the §103(a) rejection of claim 4 should be reversed.

IX. Conclusion

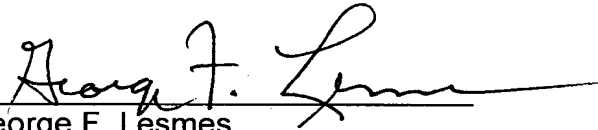
In view of the above arguments and attached Declaration, it is respectfully submitted that the §103(a) rejections of claims 1, 2, and 4-17 are unsound and should be reversed. Such action is earnestly solicited.

Respectfully submitted,

Burns, Doane, Swecker & Mathis, L.L.P.

Date March 29, 2004

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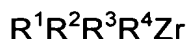
## **APPENDIX A**

### **The Appealed Claims**

1. A propylene polymer composition which is the product obtained by the steps comprising:

polymerizing propylene in the presence of an olefin polymerization catalyst comprising

(i) (a) a zirconocene compound represented by the following formula



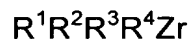
wherein two of  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  are each a substituted indenyl group substituted with aryl group, and linked together through a dimethylsilylene; and remaining two of  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  are each a halogen atom, and

(ii) at least one organoaluminum oxy-compound,

to prepare a propylene polymer (A1) having a melt flow rate (MFR), as measured according to ASTM D-1238, at 230°C under a load of 2.16 kg, of 0.01 to 30 g/10 min.; and a molecular weight distribution (Mw/Mn), as measured by gel permeation chromatography (GPC), of 2 to 3;

polymerizing propylene in the presence of an olefin polymerization catalyst comprising

(i) (a) a zirconocene compound represented by the following formula



wherein two of  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  are each substituted indenyl group substituted with aryl group, and linked together through a dimethylsilylene; and the remaining two of  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  are each a halogen atom, and

(ii) at least one organoaluminum oxy-compound,

to prepare a propylene polymer (A2) having a melt flow rate (MFR), as measured according to ASTM D-1238, at 230°C under a load of 2.16 kg, of 30 to 1000 g/10 min.; and a molecular weight distribution (Mw/Mn), as measured by gel permeation chromatography (GPC), of 2 to 4; wherein the ratio ((A2)/(A1)) of the

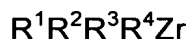
MFR of said propylene polymer (A2) to the MFR of said propylene polymer (A1) is not less than 30; and

mixing 10 to 90% by weight of the propylene polymer (A1) and 10 to 90% by weight of the propylene polymer (A2).

2. A propylene polymer composition which is the product obtained by a multi-stage polymerization method comprising the steps of:

polymerizing propylene in the presence of an olefin polymerization catalyst comprising

(i) (a) a zirconocene compound represented by the following formula



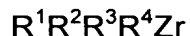
wherein two of  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  are each substituted indenyl group substituted with aryl group, and linked together through a dimethylsilylene; and the remaining two of  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  are each a halogen atom, and

(ii) at least one organoaluminum oxy-compound,

to prepare a propylene polymer (A1) having a melt flow rate (MFR), as measured according to ASTM D-1238, at 230°C under a load of 2.16 kg, of 0.01 to 30 g/10 min.; and a molecular weight distribution ( $M_w/M_n$ ), as measured by gel permeation chromatography (GPC), of 2 to 3;

polymerizing propylene in the presence of an olefin polymerization catalyst comprising

(i) (a) a zirconocene compound represented by the following formula



wherein two of  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  are each substituted indenyl group substituted with aryl group, and linked together through dimethylsilylene and the remaining two of  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  are each a halogen atom, and

(ii) at least one organoaluminum oxy-compound,

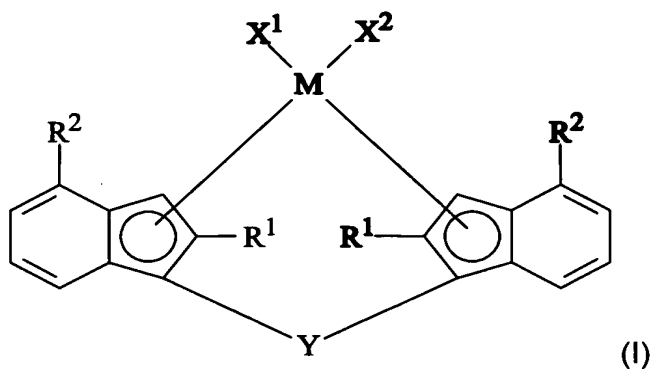
to prepare a propylene polymer (A2) having a melt flow rate (MFR), as measured according to ASTM D-1238, at 230°C under a load of 2.16 kg, of 30 to 1000 g/10 min.; and a molecular weight distribution (Mw/Mn), as measured by gel permeation chromatography (GPC), of 2 to 4; wherein the ratio ((A2)/A1)) of the MFR of said propylene polymer (A2) to the MFR of said propylene polymer (A1) is not less than 30; and

wherein the steps of preparing the propylene polymers (A1) and (A2) are conducted in an arbitrary order; and the amount of the propylene polymer (A1) is 10 to 90% by weight, the amount of the propylene polymer (A2) is 10 to 90% by weight.

4. The propylene polymer composition as claimed in claim 1 or 2, which further comprises, blended therewith, 3 to 30 parts by weight, based on 100 parts by weight of total amount of propylene polymers (A1) and (A2), of a soft polymer (B) which is a (co)polymer of ethylene or an  $\alpha$ -olefin of 3 to 20 carbon atoms, and having MFR, as measured at 190°C under a load of 2.16 kg, of 0.01 to 100 g/10 min., and a crystallinity, as measured by x-ray diffractometry, of less than 30%.

5. The propylene polymer composition as claimed in claim 3, which further comprises, blended therewith, 3 to 30 parts by weight, based on 100 parts by weight of total amount of propylene polymers (A3) and (A2), of a soft polymer (B) which is a (co)polymer of ethylene or an  $\alpha$ -olefin of 3 to 20 carbon atoms, and having MFR, as measured at 190°C under a load of 2.16 kg, of 0.01 to 100 g/10 min., and a crystallinity, as measured by x-ray diffractometry, of less than 30%.

6. The propylene polymer composition according to claim 1 wherein the zirconocene compound (i)(a) used to prepare propylene polymer (A1) and propylene polymer (A2) is a compound represented by the formula (I):



wherein M represents a zirconium atom;

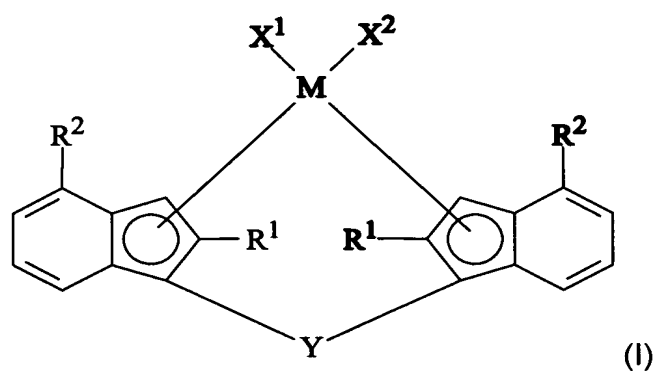
X<sup>1</sup> and X<sup>2</sup> each represent a halogen atom;

R<sup>1</sup> represents an alkyl group of from 2 to 6 carbon atoms;

R<sup>2</sup> represents an aryl group having from 6 to 16 carbon atoms; and

Y represents dimethylsilylene.

7. The propylene polymer composition according to claim 2 wherein the zirconocene compound (i)(a) used to prepare propylene polymer (A1) and propylene polymer (A2) is a compound represented by the formula (I):



wherein M represents a zirconium atom;

X<sup>1</sup> and X<sup>2</sup> each represent a halogen atom;

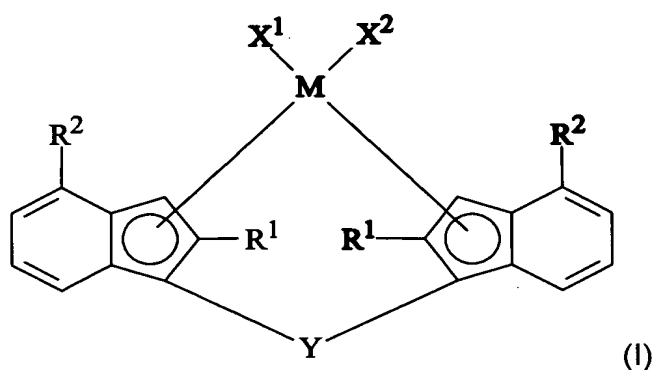
R<sup>1</sup> represents an alkyl group of from 2 to 6 carbon atoms;



$R^2$  represents an aryl group having from 6 to 16 carbon atoms; and

Y represents dimethylsilylene.

8. The propylene polymer composition according to claim 3 wherein the zirconocene compound (i)(a) is a compound represented by the formula (I):



wherein M represents a zirconium atom;

$X^1$  and  $X^2$  each represent a halogen atom;

$R^1$  represents an alkyl group of from 2 to 6 carbon atoms;

$R^2$  represents an aryl group having from 6 to 16 carbon atoms; and

Y represents dimethylsilylene.

9. The propylene polymer composition according to claim 1 wherein the zirconocene compound (i)(a) used to prepare propylene polymer (A1) and propylene polymer (A2) is rac-dimethylsilyl-bis(2-ethyl-4-phenylindenyl)zirconium dichloride.

10. The propylene polymer composition according to claim 2 wherein the zirconocene compound (i)(a) used to prepare propylene polymer (A1) and propylene polymer (A2) is rac-dimethylsilyl-bis(2-ethyl-4-phenylindenyl)zirconium dichloride.

11. The propylene polymer composition according to claim 3 wherein the zirconocene compound (i)(a) is rac-dimethylsilyl-bis(2-ethyl-4-phenylindenyl)zirconium dichloride.

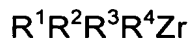
12. A propylene polymer composition comprising a physical or chemical blended mixture of from 10 to 90% by weight of first propylene polymer (A1) and from 10 to 90% by weight of second propylene polymer (A2),

wherein polymer (A1) has a melt flow rate (MFR), measured according to ASTM D-1238, at 230°C, under a load of 2.16 kg, of 0.01 to 30 g/10min; and a molecular weight distribution (Mw/Mn), measured by gel permeation chromatography (GPC), of 2 to 3; and

wherein propylene polymer (A2) has a melt flow rate (MFR), measured according to ASTM D-1238, at 230°C, under a load of 2.16 kg, of 30 to 1000 g/10 min; and a molecular weight distribution (Mw/Mn), measured by gel permeation chromatography (GPC), of 2 to 4; and

wherein propylene polymer (A1) and propylene polymer (A2) are each obtained by polymerizing propylene in the presence of an olefin polymerization catalyst comprising

(i)(a) a zirconocene compound represented by the formula



wherein  $R^1$  and  $R^2$  each represent indenyl substituted with an alkyl group and an aryl group;

$R^3$  and  $R^4$  each represent a halogen atom;

and wherein the two substituted indenyl groups are linked to each other through dimethylsilylene; and

(ii) at least one organoaluminum oxy-compound; and

wherein the ratio of the MFR of propylene polymer (A2) to the MFR of propylene polymer (A1) is not less than 30.

13. The propylene polymer composition according to claim 12 wherein propylene polymer (A1) has a crystallinity of not less than 40% and contains not more than 10 mole% of another olefin; and

propylene polymer (A2) has a crystallinity of not less than 40% and contains not more than 5 mole% of another olefin.

14. The propylene polymer composition according to claim 13 wherein propylene polymer (A1) is a propylene homopolymer.

15. The propylene polymer composition according to claim 14 wherein propylene polymer (A2) is propylene homopolymer.

16. The propylene polymer composition according to claim 13 wherein propylene polymer (A2) is a propylene homopolymer.

17. The propylene polymer composition according to claim 13 which comprises from 30 to 70% by weight of propylene polymer (A1) and from 30 to 70% by weight of propylene polymer (A2).

## **APPENDIX B**

3. A propylene polymer composition which is the product obtained by the steps comprising:

polymerizing propylene in the presence of an olefin polymerization catalyst comprising

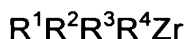
(d) a solid titanium catalyst compound, and

(e) an organoaluminum compound catalyst component,

to prepare a propylene polymer (A3) having a melt flow rate (MFR), as measured according to ASTM D-1238, at 230°C under a load of 2.16 kg, of 0.01 to 30 g/10 min.; and a molecular weight distribution (Mw/Mn), as measured by gel permeation chromatography (GFC), of 4 to 15;

polymerizing propylene in the presence of an olefin polymerization catalyst comprising

(i)(a) a zirconocene compound represented by the following formula



Wherein two of  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  are each a substituted indenyl group substituted with aryl group, and linked together through dimethylsilylene and the remaining two of  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  are each a halogen atom, and

(ii) at least one organoaluminum oxy-compound,

to prepare a propylene polymer (A2) having a melt flow rate (MFR), as measured according to ASTM D-1238, at 230°C under a load of 2.16 kg, of 30 to 1000 g/10 min.; and a molecular weight distribution (Mw/Mn), as measured by gel permeation chromatography (GPC), of 2 to 4; and

mixing 10 to 90% by weight of propylene polymer (A3) and 10 to 90% by weight of the propylene polymer (A2).

COPY

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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In re Application of  
Mikio HASHIMOTO, et al.  
Serial No. 08/490,608  
Filed: June 7, 1995

Group Art Unit: 1713  
Examiner: D.R. Wilson

For: PROPYLENE POLYMER COMPOSITIONS

The Honorable Commissioner of Patents and Trademarks  
United States Patent and Trademark Office  
Washington, D. C. 20231

Sir:

DECLARATION UNDER 37 CFR 1.132

I, Ryoji MORI, declare and state that:

1. I am a citizen of Japan, residing at 1-2, Muronoki-cho, Iwakuni-shi, Yamaguchi, Japan.

In March, 1990, I was graduated from Kyushu Institute of technology, Industrial chemistry of the faculty of Engineering, and received a Bachelor of Engineering from the same university. In March, 1992, I was graduated from Molecular Science and technology of the interdisciplinary Graduated School of Engineering Science, Kyushu University, and received a Master of Engineering from the same University.

Since April, 1992, I have been an employee of MITSUI Petrochemical Industries Ltd., and till the present time, I have been engaged in research and development work in Polymer Science.

2. I am a researcher of MITSUI Petrochemical Industries Ltd. (now, MITSUI CHEMICALS INC.), who is an assignee of the present application.

3. The following Experiments were carried out in order to demonstrate the superiority of the presently claimed composition.

Catalyst system substituent on indenyl ring	PP	MFR g/10 min	Exp. 1	Exp. 2	Comp Exp. 1	Comp Exp. 2	Comp Exp. 3	Comp Exp. 4	Ref. Exp. 1	Ref. Exp. 2
2-Et-4-Ph	(7)	900	70	30						
2-Et-4-Ph	(8)	0.75	30	70					100	
2-Me-4- <sup>i</sup> Pr	*1	2.3			70	30				
2-Me-4- <sup>i</sup> Pr	*2	610			30	70				
2-Me-4-Ph	*3	1.3								100
Ti-catalyst	*4	490					70	30		
Ti-catalyst	(6)	25					30	70		
HDT (°C)			123	121	108	109	119	116	118	116

Composition of Experiment 1 comprises 70 parts by weight of PP(7) and 30 parts by weight of PP(8).  
PP(6)-(8) are the same as those obtained in the present specification.

\*1: Polymerization condition was modified so as obtain PP-homopolymer having MFR of 2.3 g/10min.

\*2: Polymerization condition was modified so as obtain PP-homopolymer having MFR of 610 g/10min.

\*3: Polymerization condition was modified so as obtain PP-homopolymer having MFR of 1.3 g/10min.

\*4: Procedure of preparation PP(6) of the present specification was repeated, except for modifying polymerization condition so as obtain PP-homopolymer having MFR of 410 g/10min.

4. From the results of the above experiments compared with the Examples of the present application, and based on my knowledge and experience on Polymer Chemistry, I conclude that:

PP compositions comprising two different PPs both obtained by the specific metallocene catalyst containing

aryl-substituted indenyl ligands realize remarkable improvement regarding HDT.

The undersigned declares further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Respectfully submitted,

*Ryoji Mori*

this 12<sup>th</sup> day of July, 2000

- Ryoji Mori